Supporting materials

Table S1 Results of slab thickness convergence tests

Surface	Layer thicknesse	E_S/eV	n	E_B/eV	A/Å^2	γ/(eV·Å^{-2})
Illite(001)	1	−300.139	40	−7.603	46.269	0.043
	2	−595.574	80	−7.603	46.269	0.137
	3	−906.731	120	−7.603	46.269	0.061
	4	−1210.968	160	−7.603	46.269	0.060
Na-MMT(001)	1	−298.020	41	−7.299	47.771	0.001
	2	−596.816	82	−7.299	47.771	0.018
	3	−895.717	123	−7.299	47.771	0.022
	4	−1194.675	164	−7.299	47.771	0.025

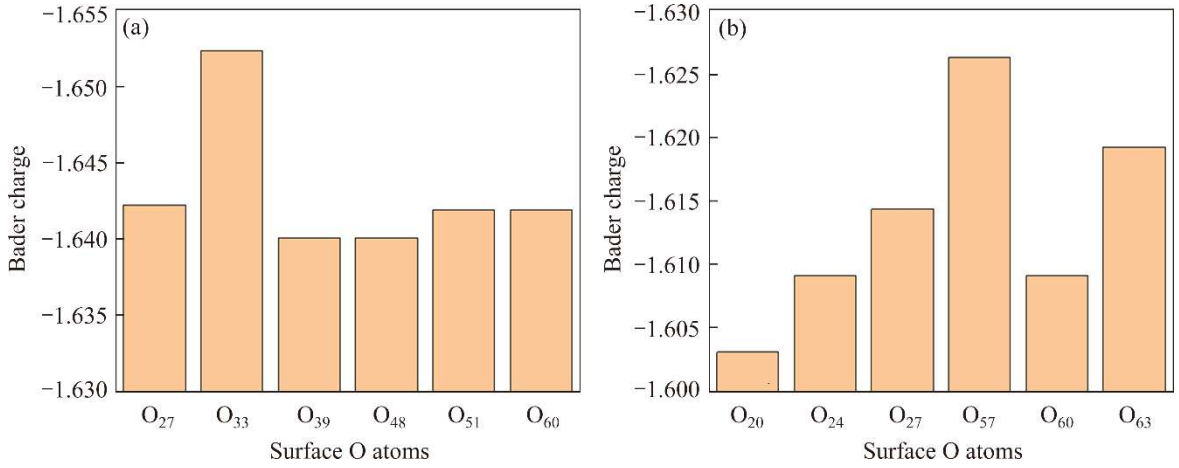

Figure S1 Bader charge of surface O atoms: (a) Illite(001) surface; (b) Na-MMT(001) surface

Table S2 Recovery time of adsorbed surface

Surface	Adsorbate	E_{ad}/eV	τ/s
Illite(001)	As atom	−1.94	6.45×10^{19}
	H_3AsO_3 molecule	−1.40	4.75×10^{10}
Na-MMT(001)	As atom	−0.56	2.96×10^{-4}
	H_3AsO_3 molecule	−1.01	1.21×10^{4}

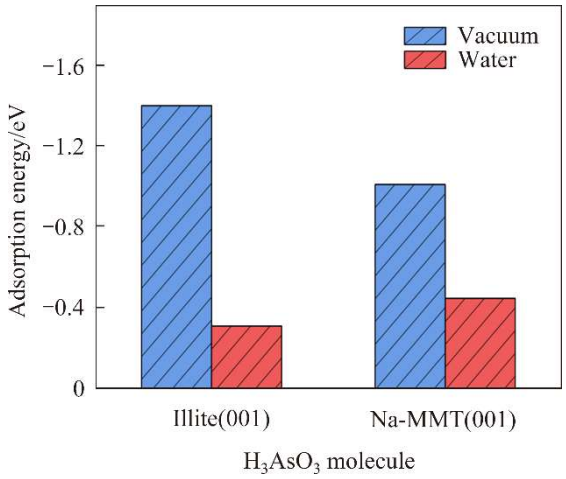

Figure S2 Comparison of adsorption energy and solvated adsorption energy of H_3AsO_3 molecules on illite(001) and Na-MMT(001) surfaces

Table S3 The adsorption energies of the Hg, Cd and Cr atoms on Na-MMT(001) surfaces

Type	Adsorption sites	Adsorption energy/eV
Hg	Top(Na_3)	−0.07
	Top(O_{20})	−0.02
	Top(O_{24})	−0.03
	Hollow	−0.02
Cd	Top(Na_3)	−0.06
	Top(O_{20})	−0.03
	Top(O_{24})	−0.04
	Hollow	−0.03
Cr	Top(Na_3)	−0.17
	Top(O_{20})	−0.06
	Top(O_{24})	−0.16
	Hollow	−0.06

Table S4 Valence electron of O atoms of H_3AsO_3 molecule before and after adsorption on illite(001) and Na-MMT(001) surfaces

Surface	H_3AsO_3 molecule (Before adsorption)			H_3AsO_3 molecule (After adsorption)		
	O_{73}	O_{74}	O_{75}	O_{73}	O_{74}	O_{75}
Illite(001)	7.13	7.15	7.15	7.22	7.17	7.17
Na-MMT(001)				7.16	7.17	7.16

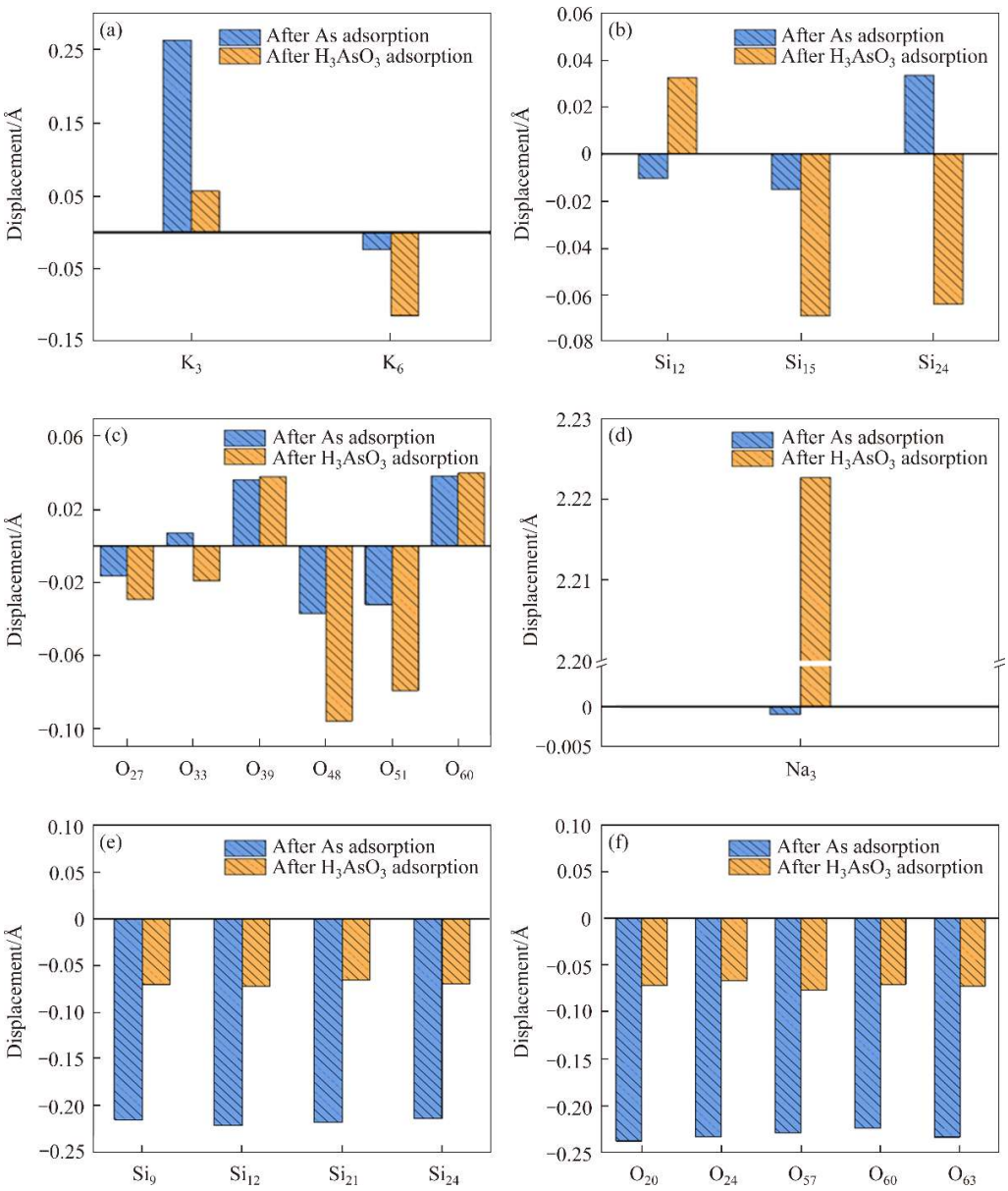

Figure S3 *Z*-direction displacements of surface atoms in the surface: Surface K atoms (a), surface Si atoms (b), and surface O atoms (c) of illite(001); Surface Na atoms (d), surface Si atoms (e) and surface O atoms (f) of Na-MMT(001) (Positive displacements represent movement toward the bulk exterior)

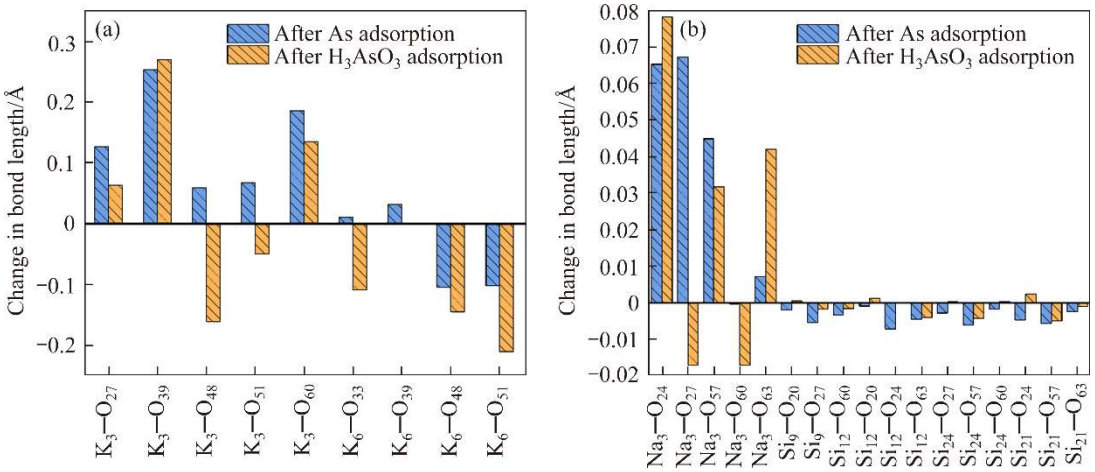

Figure S4 Changes in bond lengths of surface atoms: (a) Illite(001); (b) Na-MMT(001) (Positive value represent the increase in bond length)

Table S5 Valence electron of surface O atom before and after adsorption on illite(001) and Na-MMT(001) surfaces

Surface	Atom	Before adsorption	After As atom adsorption	After H_3AsO_3 molecule adsorption
Illite(001)	O_{51}	7.64	7.61	7.61
	O_{60}	7.64	7.61	7.61
	O_{39}	7.64	7.61	7.60
	O_{48}	7.64	7.61	7.60
	O_{33}	7.65	7.62	7.62
	O_{27}	7.64	7.62	7.61
	O_{69}	7.58	7.58	7.58
	O_{15}	7.59	7.60	7.60
	O_{24}	7.59	7.60	7.60
	O_3	7.61	7.60	7.45
	O_{12}	7.61	7.43	7.59
	O_{63}	7.43	7.61	7.61
	O_{66}	7.42	7.61	7.61
	O_6	7.61	7.42	7.60
	O_9	7.61	7.60	7.41
	O_{18}	7.60	7.61	7.61
	O_{21}	7.60	7.60	7.61
	O_{72}	7.57	7.58	7.58
Na-MMT(001)	O_{24}	7.60	7.60	7.60
	O_{27}	7.61	7.60	7.60
	O_{63}	7.61	7.60	7.60
	O_{60}	7.60	7.61	7.61
	O_{20}	7.60	7.60	7.60
	O_{57}	7.62	7.62	7.62
	O_{30}	7.41	7.42	7.42
	O_{33}	7.60	7.60	7.60
	O_{72}	7.60	7.60	7.60
	O_{36}	7.60	7.61	7.60
	O_{69}	7.61	7.60	7.61
	O_{64}	7.41	7.43	7.43
	O_{11}	7.42	7.44	7.45
	O_{48}	7.43	7.61	7.62
	O_{15}	7.61	7.43	7.43
	O_{54}	7.62	7.61	7.61
	O_{18}	7.61	7.61	7.61
	O_{50}	7.61	7.60	7.61

Table S6 Valence electron of surface Si atom before and after adsorption on illite(001) and Na-MMT(001) surfaces

Surface	Atom	Before adsorption	After As atom adsorption	After H_3AsO_3 molecule adsorption
Illite(001)	Si_{15}	0.84	0.83	0.83
	Si_{24}	0.84	0.83	0.83
	Si_3	0.83	0.83	0.83
	Si_{12}	0.83	0.82	0.82
Na-MMT(001)	Si_{12}	0.84	0.83	0.83
	Si_9	0.83	0.82	0.83
	Si_{21}	0.84	0.83	0.84
	Si_{24}	0.83	0.83	0.82

Table S7 Valence electron of As atom and H_3AsO_3 molecule before and after adsorption on illite(001) and Na-MMT(001) surfaces

Surface	As atom (Before adsorption)	As atom (After adsorption)	H_3AsO_3 molecule (Before adsorption)	H_3AsO_3 molecule (After adsorption)
Illite(001)	5.00	5.71	26.00	26.74
Na-MMT(001)	5.00	5.13	26.00	26.06

Table S8 Valence electron transfer of surface Na atom and adjacent surface O atoms after adsorption on Na-MMT(001) surfaces. Note that the negative value represents the loss of the electron

Atom	After As atom adsorption	After H_3AsO_3 molecule adsorption
Na	0.005	0.002
O_{24}	−0.001	−0.005
O_{27}	−0.008	−0.008
O_{57}	−0.002	−0.006
O_{60}	−0.012	−0.010
O_{63}	0.003	0.004

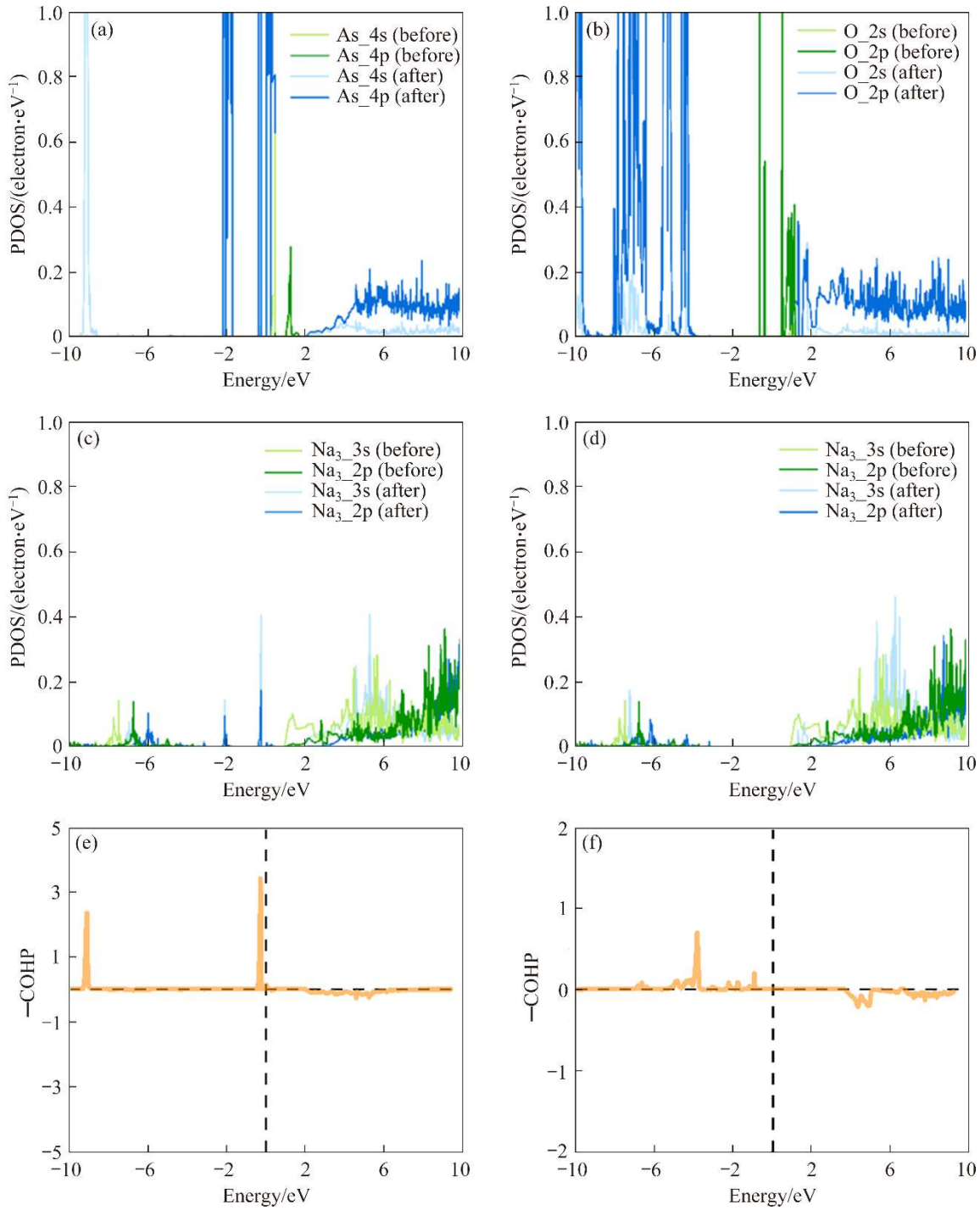

Figure S5 PDOS and —COHP of Na-MMT(001) before and after As atom and H_3AsO_3 molecule adsorption: (a) As atom; (b) O atoms of H_3AsO_3 molecule; (c) Surface Na_3 atom before and after As atom adsorption; (d) Surface Na_3 atom before and after H_3AsO_3 molecule adsorption; (e) As—Na bond in As atom adsorption; (f) O—Na bond in H_3AsO_3 molecule adsorption (The Fermi energy level was set as 0 eV)